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14. ABSTRACT
The goals of this program were to develop optical coatings that can resist or circumvent the detrimental effects of thermal gradients. New thin film materials were investigated and optomechanical design techniques were developed. Over the three years of the program, we have made major progress on three main thrusts. These are: 1) encapsulation techniques to stabilize the optical and mechanical properties of porous, reactive, or unstable thin films, 2) optomechanical design to create optical coatings with specified thermal and mechanical deformation, 3) negative thermal expansion (NTE) films that are stable with changing atmospheres. In addition we have preliminary results regarding two additional areas of inquiry that could have great impact on the coatings for high power lasers. These are: 4) infrared signatures to predict the onset of laser damage to optical coatings, and 5) atomic layer deposition for highly conformal coatings for high power beam combining.

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			**17. LIMITATION OF ABSTRACT**	**18. NUMBER OF PAGES**	**19a. NAME OF RESPONSIBLE PERSON**
a. REPORT	**b. ABSTRACT**	**c. THIS PAGE**			Joseph Talghader
U	U	U	U		**19b. TELEPHONE NUMBER** *(Include area code)* (612) 625-4524

I. Objectives:

No Change

II. Status of effort (Limit to 200 words): (A brief statement of progress towards achieving the research objectives.)

In the past year, our program has concentrated on three main thrusts:

a) Encapsulation techniques to stabilize the optical and mechanical properties of porous, reactive, or unstable thin films. The purpose of this thrust is to allow one to use materials with unusual but highly desirable properties in high energy laser optical coatings without impacting the environmental robustness of the overall stack.

Specifically, we have developed a deposition technique for nanoporous silica (np-SiO_2) films. The films, which have a refractive index and coefficient of thermal expansion that vary with humidity, have been successfully encapsulated by an Al_2O_3 layer using atomic layer deposition.

b) Extreme temperature optical design to create optical coatings that are deformation free over hundreds of degrees. From the simulations, high-reflectance coating designs based on a HfO_2 and SiO_2 multilayer system have been optimized to show edge deflection < 0.1nm/K, an order of magnitude less than "standard" designs while maintaining similar reflectance performance. Experimental testing is currently in progress and should be completed in the remaining months of 2007.

c) Negative thermal expansion (NTE) films that are stable with changing atmospheres. These films have been tested up to 300 °C in air and N_2 and have been developed by sputtering ultra-low expansion (ULE) glass provided by Corning. We have found that the sputtering conditions such as RF power and Ar pressure and flow rate lead to the change in CTE of films.

III. Accomplishments/New Findings: (Describe research highlights, their significance to the field, their relationship to the original goals, their relevance to the AF's mission, and their potential applications to AF and civilian technology challenges.)

The critical new findings of the last 12 months the program are:

1) Al_2O_3 thin films deposited by atomic layer deposition (ALD) can be used to encapsulation of porous oxide coatings. This allows one to use materials with unusual but highly desirable properties in high energy laser optical coatings without impacting the environmental robustness of the overall stack.

2) Simulations based on measured material parameters indicate that HfO_2 and SiO_2 films can be used to design high reflectance coatings optimized to be thermally invariant over a wide temperature range.

3) A new negative thermal expansion material based on ultra-low expansion glass (ULE) (Corning 7972) has been developed under certain sputtering conditions. This film has a stable thermal expansion coefficient in both air and dry nitrogen between 20°C and 300°C. Further studies on co-sputtered SiO_2 and TiO_2 are in progress to optimize this material for HEL applications.

1) Encapsulation to stabilize reactive optical coatings such as np-SiO_2 using sputtering and atomic layer deposition (ALD) methods:

np-SiO_2 thin films with various refractive indices and CTEs have been deposited on (100) Si substrates using e-beam evaporation. The index of refraction of np-SiO_2 can be controlled by changing process conditions, namely the substrate angle relative to the source, the deposition rate, pressure, *etc.* This can be used to enhance the performance of a standard DBR, or it can enable more exotic solutions, such as fabrication of a DBR using a single source material. However, typical np-SiO_2 has material properties that depend on environmental conditions, especially humidity. In order to make mechanically stable coatings without sacrificing desirable optical properties, proper encapsulation of this np-SiO_2 is essential. For encapsulation using sputtering, Al_2O_3 was deposited on top of np-SiO_2 using an Al_2O_3 target with a RF magnetron sputter system. The radius curvature (1/radius) of the Al_2O_3/np-SiO_2/Si system was measured in air and dry N_2 over a temperature range of 20 °C to 100 °C using an optical film stress measurement instrument. From these data, the CTE of np-SiO_2 was calculated using a multilayer free-plate model. The derived CTE of the encapsulated np-SiO_2 in air and N_2 were 6.35 and 6.27 ppm/K, respectively, as shown in Fig. 1-(1).

Encapsulation using ALD Al_2O_3 has also been demonstrated, with results shown in Fig. 1-(2). This Al_2O_3 layer was deposited on top of np-SiO_2 at a process temperature of 250°C using a Cambridge NanoTech Inc. Savannah™ ALD system. Thermomechanical characterization has been performed in the same way as the sputtered Al_2O_3/np-SiO_2/Si. The CTE of the encapsulated np-SiO_2 in air and N_2 were 6.12 and 6.21 ppm/K, respectively.

Fig. 1-(3) shows a cross-sectional SEM image of the np-SiO_2/Si; evaporated np-SiO_2 tends to have a columnar structure which leads to reduced density and index of refraction.

We have also characterized the temperature dependence of the refractive index of np-SiO_2 using ellipsometry at the temperature range of 10 to 65 °C, as shown in Fig. 1-(4). Note that this result appears to be dominated by the effect of adsorbed water vapor in the pores of the film since it is done in an ambient with 40% relative humidity.

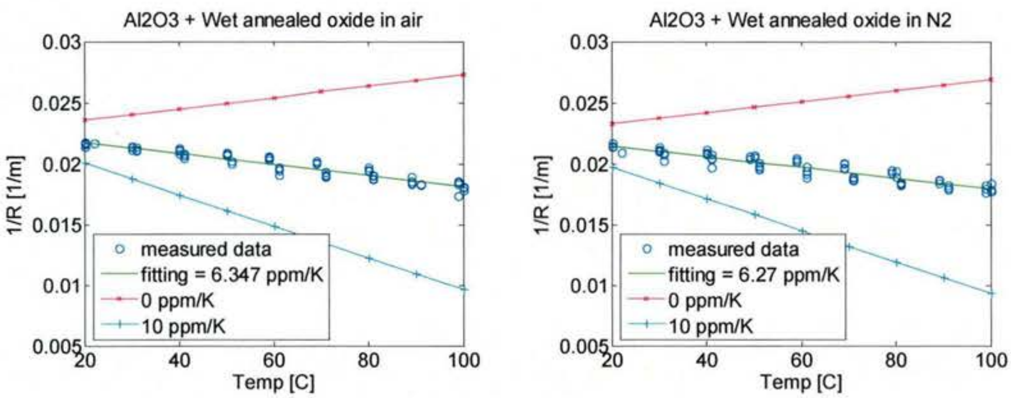

Fig. 1-(1): CTE of np-SiO_2 when encapsulated by sputtered Al_2O_3; measured in air (left) and dry N_2 (right).

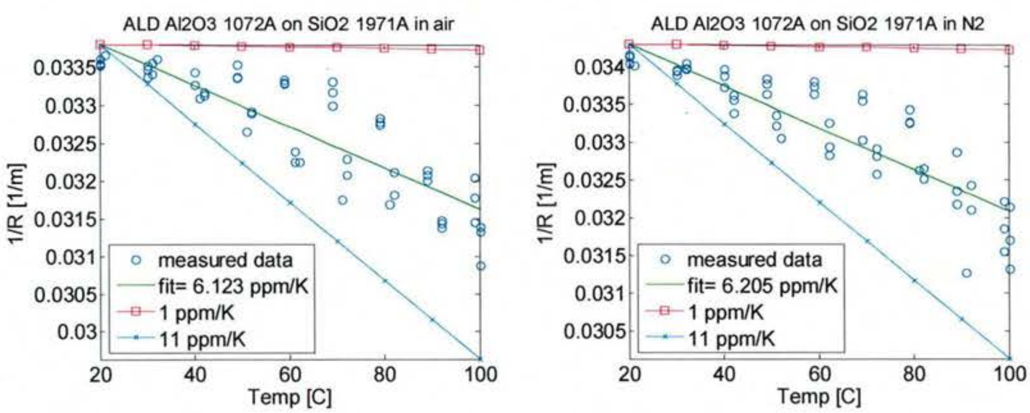

Fig. 1-(2): Measured CTE np-SiO_2 when encapsulated by Al_2O_3 formed by ALD; measured in air (left) and dry N_2 (right).

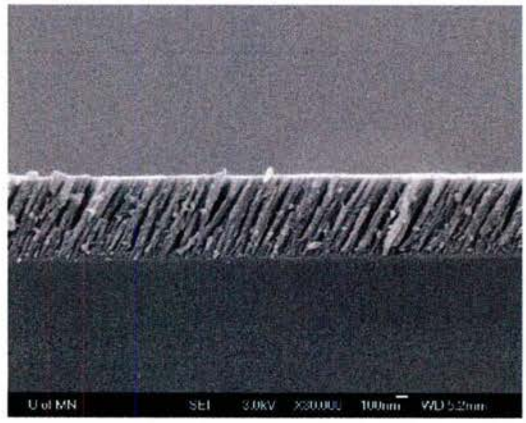

Fig. 1-(3): SEM image of the cross-section of np-SiO_2 on a Si wafer.

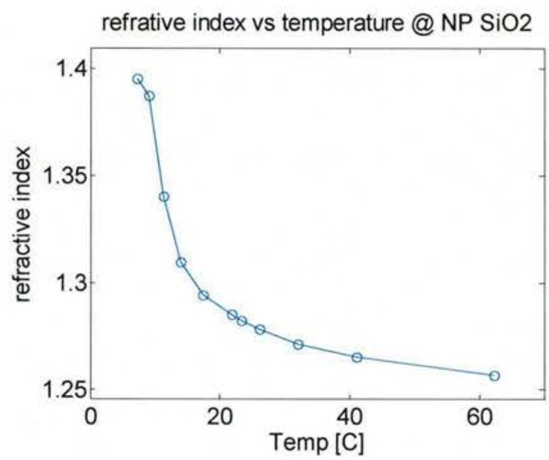

Fig. 1-(4): The temperature dependence of the refractive index of np-SiO_2.

2) <u>Extreme temperature optical design to create optical coatings that are deformation free over hundreds of degrees.</u>

i) High-reflectance, low-absorption mirrors: The primary way to alleviate thermomechanical deformation of a micromirror under illumination is to increase the reflectance of the mirror. A multi-layer dielectric mirror becomes a necessity at moderate to high energies because it can allow near 100% reflectance to be achieved. This implies near-zero absorption of optical power and less heating of the mirror structure.

This technique was successfully demonstrated on a micromirror array fabricated by Sandia National Laboratory. Fig. 2-(1) shows the curvature of a micromirror roughly 500 μm in diameter under varying laser illumination, with and without a 3-pair DBR mirror designed and fabricated by our group. As is clear from these data, the addition of the dielectric mirror greatly reduced thermal deformation, with peak applied optical power density over 2.5kW/cm^2. Note that no special attention was paid in this design to prevent thermal deformation of the structure due to CTE mismatch; significantly increasing the reflectance above that of the base AlCu layer was the design goal. We gratefully acknowledge the collaborative efforts of Dr. Olga Blum Spahn in this experiment.

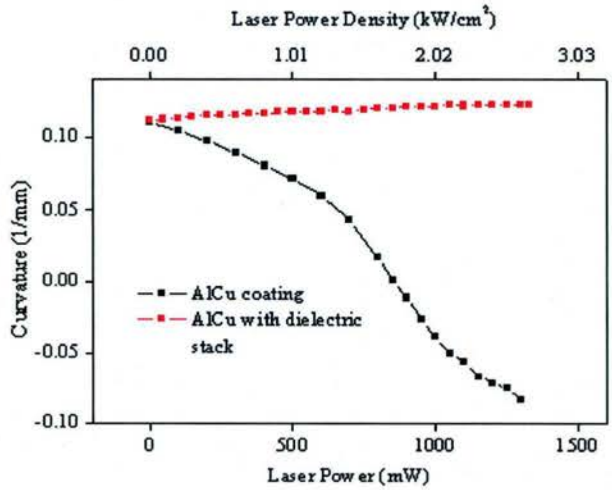

Fig. 2-(1): Micromirror curvature vs. laser illumination power with and without a DBR mirror coating. Microstructures and experimental data courtesy of Dr. Olga Blum Spahn, Sandia National Laboratories.

ii) Reduction of thermal deformation: Since 100% reflectance, *i.e.* zero absorption, is not practically achievable, for very high energy lasers a secondary method of reducing thermal deformation is needed. This can be done by balance thermal stresses through appropriate choice of thermal expansion coefficient (CTE), modulus, and thickness of each constituent layer of a dielectric mirror. Since the effect of varying modulus is relatively small and difficult to achieve in practice, our focus is on the CTE and thickness of each layer. For our design, at least one layer of the dielectric mirror must have a CTE less than that of the base structural material, and at least one layer must have a CTE greater than the structural material. This is because the relative signs of the "CTE mismatch" determine the direction of thermal curvature, and the goal here is to balance the "positive" and "negative" thermal curvature to have them essentially sum to zero. In addition to these basic requirements, the film stress and CTE must also be invariant under all other conditions, most notably ambient levels of water vapor.

We have fabricated individual films that meet the basic requirements; properly sputtered SiO_2 has a low refractive index and CTE always less than Si, and our HfO_2 films deposited by sputtering and ALD techniques have high refractive index and CTE always higher than Si. Further refinement would improve stability and predictability of CTE to make thermal deformation modeling more accurate and reproducible. The sputtered films are particularily unpredictable and have some residual variability with environmental.conditions.

iii) Optimization of optical and thermomechanical performance of SiO_2 and HfO_2 multilayer systems – zero edge-deflection: Our approach for optimization is based on numerical methods, because of the relative complexity of the mathematics involved as the number of layers increase. This is especially useful because the approach used here is to optimize all layers simultaneously, as opposed to previous efforts that used only a single "compensation layer" with varying thickness[1]. A linear free-plate model is used to simulate thermal deformation, based on the multilayer thermostat analysis by M. Vasudevan and W. Johnson[2]. This model is designed for unconstrained systems, and can be empirically adapted to simulate constrained systems (*e.g.* a micromirror with support beams.) Optical performance versus wavelength is simulated with a model based on iterative calculation of the reflection and transmission as light at a given wavelength passes through successive layers of a mirror. The optimization itself is a Nelder-Mead simplex method executed in MATLAB that attempts to minimize a custom merit function. This merit function contains terms that represent various optical and thermal performance metrics derived from the simulation, including peak reflectance, center wavelength, full-width at half maximum (FWHM), and total thermal deformation. Balancing the various terms with weighting factors allows the performance compromise to be adjusted to achieve the desired end result.

The result of this multiphysics optimization scheme is small (~ 10-100Å) variations in layer thicknesses from their starting values to greatly reduce thermal deformation while minimally affecting the optical performance. In this report, two optimized simulation results with 8-pair HfO_2/SiO_2 DBR coatings are included to indicate both

[1] W. Liu and J.J. Talghader, "Thermally invariant dielectric coatings for micromirrors", Appl. Opt. **41**, 3285-3293 (2002).

[2] "On multi-metal thermostats", Appl. Sci. Res. Sect. B **9**, pp. 420-430 (1962).

macro- and micro-scale performance for high-reflectance dielectric mirror coatings: Figure 2-(2) shows the reflectance versus wavelength and curvature versus temperature for an 8-pair SiO_2/HfO_2 DBR coating on a 100µm-thick 100mm silicon wafer. The parameters used are summarized in Table I. Figure 2-(3) shows the reflectance versus wavelength and curvature versus temperature for a similar DBR coating, this time on a 1µm thick silicon micromirror that is 500µm in diameter. The parameters used in this case are summarized in Table II. We see that in both size regimes the coating can be optimized quite effectively to eliminate thermal deformation.

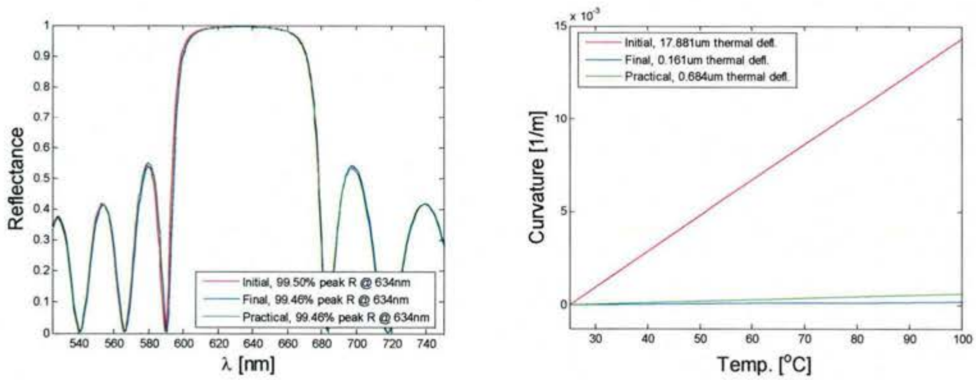

Fig. 2-(2): Simulated reflectance versus wavelength and curvature versus temperature for 8-pair SiO_2/HfO_2 DBR coating on a silicon wafer, before and after optimization.

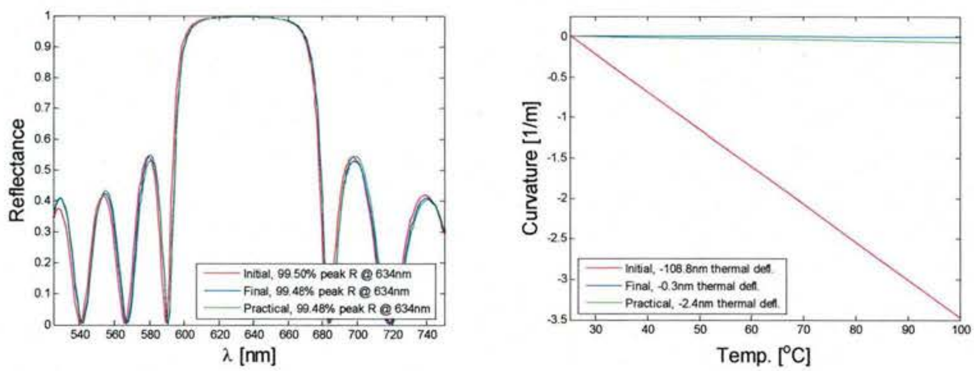

Fig. 2-(3): Simulated reflectance versus wavelength (left) and curvature versus temperature (right) for an 8-pair SiO_2/HfO_2 DBR coating on a silicon micromirror, before and after optimization.

The optimized performance above was obtained by varying the thickness of each layer on the order of 20-120Å from the nominal quarter-wave configuration. The required thickness changes in the HfO_2 layers were between 60 and 90 Å, and in the SiO_2 layers between -60 and -120Å. Layer thickness changes were needed in both directions on the micromirror, with the magnitude of HfO_2 layer variations between 20 and 70Å, and SiO_2 variations between 20 and 100Å.

3) <u>Negative thermal expansion (NTE) coating development over wide temperature range independent of ambient conditions:</u>

So far, it has been reported that oxide coatings with lower density typically show negative thermal expansion in air, but not in N_2 due to the structural changes from water vapor desorption near the surface. In our previous research, zirconium tungstate films evaporated by e-beam did show NTE behavior in air and N_2, but only over a very limited temperature range. Typically, sputtered films have higher density than evaporated films. Due to this higher density, it was previously believed that it may be difficult to obtain NTE coatings using sputtering, and positive thermal expansion was indeed always observed with sputtered zirconium tungstate. But from our recent work with ultra low expansion (ULE) glass from Corning, we have found that it is possible to sputter NTE thin films using certain process conditions (RF power, pressure, and Ar flow rate). The CTE of sputtered films in air and N_2 have been obtained from the same film stress measurement method mentioned above. The sputtered NTE films using ULE glass tend to be more stable in the presence of humidity than zirconium tungstate films. Figure 3-(1) shows the relationship of curvature versus temperature, with the CTE extracted from the slope. Table III is a summary of the process conditions for sputtering and CTE values obtained from the simulation. For NTE glass films, the best results of CTE in air and N_2 in the temperature range from 20 to 300 °C we obtained so far were -4.1 ppm/K and -1.0 ppm/K, respectively. This result was obtained under the following sputtering conditions using AJA Sputter System: 1) RF power: 250W; 2) Ar flow rate: 20 sccm; 3) Pressure: 5 mTorr; 4) Deposition time: 120 min; and 5) Thickness: 77.2 nm. Table III shows the summary of various process conditions used for sputtering and CTE values obtained from the simulation. Fig. 3-(2) shows the CTE values versus Ar pressure, with a constant RF power of 250 W. As can be seen, NTE films were obtained with the certain conditions. In addition to thermal expansion, other mechanical properties would affect high power optical coatings. One of important mechanical properties is elastic modulus. We obtained the elastic modulus of one of our NTE glass films using nanoindentation as shown in Fig. 3-(3). The measured elastic modulus of the NTE glass film was ~ 70 GPa.

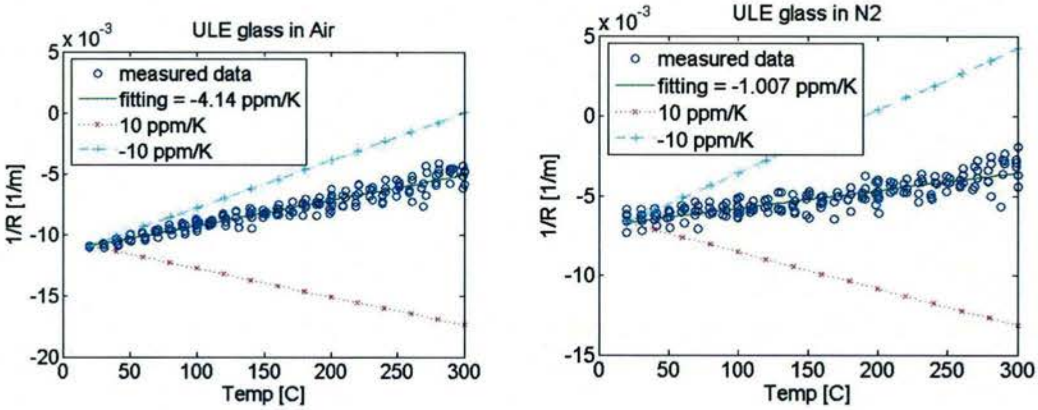

Fig. 3-(1): The relationship of curvature versus temperature and the CTE of sputtered film in air and dry N_2.

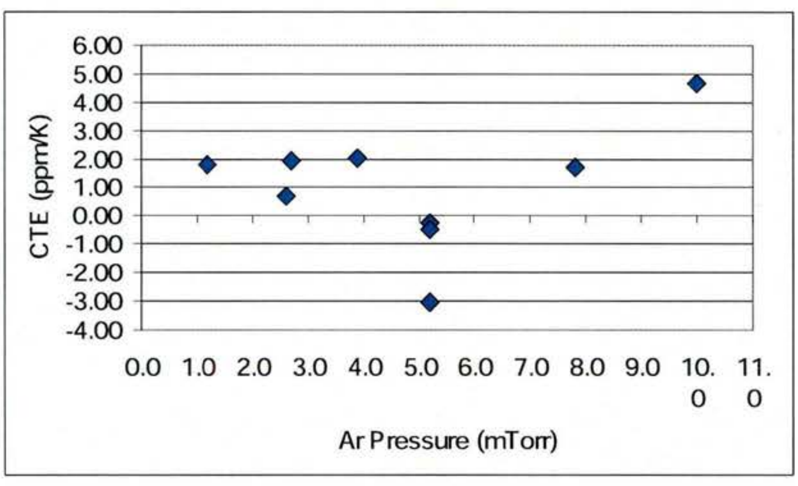

Fig. 3-(2): The CTE values of ULE glass thin films (sputtered at 250W) versus Ar pressure, measured from 20-100°C in dry N_2.

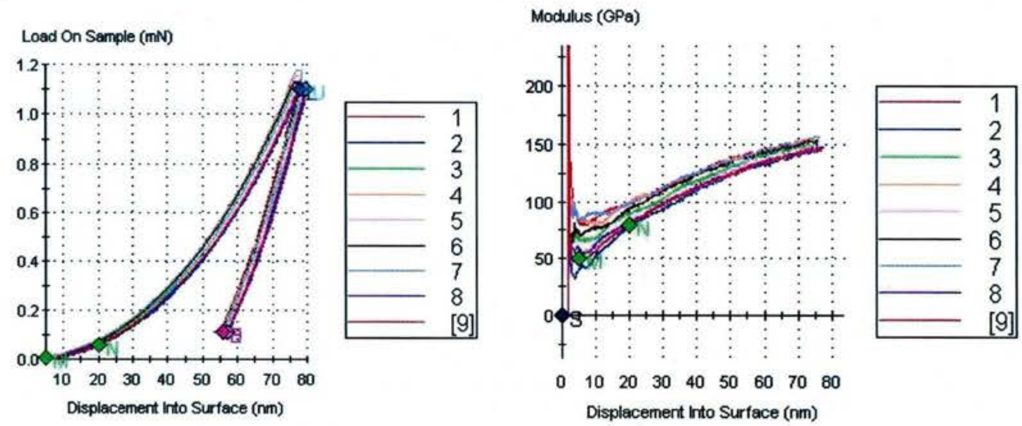

Fig. 3-(3): Nanoindentation measurement of a NTE glass film to extract the elastic modulus.

Table III: Summary of the process conditions for sputtering and associated thin film CTE values obtained from measurements wafer curvature versus temperature.

Thickness (Å)	Power (W)	Ar Flow (sccm)	O_2 Flow (sccm)	Fwd Bias Voltage (V)	Process Pressure (mTorr)	CTE (ppm/K) at 20-100C in N2	CTE (ppm/K) at 20-300C in N2	CTE (ppm/K) at 20-100C in Air
772	250	20.0	0.0	643-637	5.2E+00	-3.051	-1.007	-4.14
737	250.0	20.0	0.0	657-652	5.2E+00	- 0.2618	0.3856	-4.843
1393	350	20.0	0.0	803-793	5.2E+00	1.683		-0.8736
366	250.0	20.0	0.0	619-617	1.0E+01	4.682		
680	350	20.0	0.0	767-747	1.0E+01	2.387		
721	250.0	20.0	0.0	625-595	5.2E+00	-0.4702		
1411	250	10.0	0.0	590-580	2.6E+00	0.6987	0.0861	
730	250.0	30.0	0.0	581-579	7.8E+00	1.697		
1200	250	15.0	0.0	551-554	3.9E+00	2.007	1.317	
1403	250.0	10.0	0.0	581-557	2.7E+00		6.799	
1460	250	10.0	0.0	584-574	2.7E+00	1.949	1.835	
1989	250.0	5.0	0.0	567-546	1.2E+00	1.792	0.3446	
403	250	10.0	5.0	583-560	3.0E+00		6.211	

Significance to the field:

i) The encapsulation technique opens a new way to design opto-mechanical coatings, enabling a wider choice of materials which are typically sensitive to environmental conditions; ii) The developed oblique-angle deposition techniques can reduce the process cost to fabricate a DBR stack by allowing use of a single source material; iii) Stable thin films with negative or near-zero thermal expansion are critical to making lightweight thermally stable HEL coatings.

Relationship to the original goal:

All three current thrusts were fundamental to the original goal of developing new materials and design techniques to understand the mechanical behavior of optical coatings.

Relevance to the AF's mission:

The Air Force has a need for thermally stable and adaptive optical systems for use in high-energy laser systems. The specialty coating materials, encapsulation techniques, and design optimization methodologies developed by our group should enable a significant increase in the practical operating temperature range of the membrane-based mirrors used in adaptive optics. This is crucially important in order to maintain specified performance with high levels of laser illumination since these structures are so susceptible to heating. Stability of such structures in adverse ambient conditions can also be improved.

Their potential applications to AF and civilian technology challenges:

i) Increasing mechanical performance of optical coatings and the ability to coat optical or other membranes to high stability.
ii) Creation of thin films stacks for optical coatings and electronic packaging that are thermal expansion matched to substrates with specialized thermal expansion coefficients. This will have applications both in optics and standard integrated circuits.

IV. Personnel Supported: (List professional personnel (Faculty, Post-Docs, Graduate Students, etc.) supported by and/or associated with the research effort.)

1) Faculty: Professor Joseph J. Talghader (PI) and Professor Philip I. Cohen
2) Post-Docs: Dr. Woo-Bin Song
3) Graduate Students: Nicholas Gabriel, Sangho Kim, Ryan Shea, Andy Bettino
4) Undergraduates: Merlin Mah

V. Archival publications (published) during reporting period:

1) M. T. K. Soh, J. J. Thomas III, and J. J. Talghader, "Thermally induced structural changes in nanoporous silicon dioxide from x-ray photoelectron spectroscopy," *J. Vac. Sci. Technol. A* **24(6)**, 2147, 2006.
2) M. T. K. Soh, J. Thurn, J. J. Thomas III, and J. J. Talghader, "Thermally induced stress hysteresis and co-efficient of thermal expansion changes in nanoporous SiO_2," *J. Phys. D: Appl. Phys.* **40**, 2176, 2007.
3) S. S. Kim, N. T. Gabriel, W.-B. Song, and J. J. Talghader, "Encapsulation of low-refractive-index SiO_2 nanorods by Al_2O_3 with atomic layer deposition," submitted to *Optics Express*, September 1, 2007.

VI. Interactions/Transitions:

a. Participation/presentations at meetings, conferences, seminars, etc.:
J. J. Talghader, "From micro- to nano-: thermal effects in micromachined devices," 2006 Conference on Optoelectronic and Microelectronic Materials and Devices, Perth, Western Australia, December 6-8, 2006.

b. Consultative and advisory functions to other laboratories and agencies, especially Air Force and other DoD laboratories, Provide factual information about the subject matter, institutions, locations, dates, and name(s) of principal individuals involved:
1) Sandia National Laboratories, Albuquerque, NM, Dr. Olga Blum Spahn (See Section III, 2(a)) – An array of micromirrors capable of handling high optical powers were integral to research at Sandia. We developed a thin film stack that could simultaneous boost power-handling capacity without introducing excessive amounts of stress.
2) Northrop Grumman, John Starkovich. Over a year ago, we made a contact with Northrop Grumman to develop negative expansion coatings for space qualification testing. In the interim, we have been steadily working towards thin film materials that have negative thermal expansion in dry/space environments. We are currently refining our ULE glass-based deposition process and will be sending the Kapton membrane back to NG for testing in the coming months of 2007.
3) Interaction to measurement of thermal properties of niobia thin films for David Reicher of S Systems.

c. Transitions. Describe cases where knowledge resulting from your effort is used, or will be used, in a technology application. Transitions can be to entities in the DoD, other federal agencies, or industry. Briefly list the enabling research, the laboratory or company, and an individual in that organization who made use of your research:

1) Sandia National Laboratories, Albuquerque, NM, Dr. Olga Blum Spahn (See Section III, 2(a)) – An array of micromirrors capable of handling high optical powers were integral to research at Sandia. We developed a thin film stack that could simultaneous boost power-handling capacity without introducing excessive amounts of stress.
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VII. New discoveries, inventions, or patent disclosures. (If none, report None)
1) A new negative thermal expansion thin film has been discovered by RF sputtering ultra low expansion glass (Corning 7972). These films appear to be stable in both wet and dry ambients but they only occur under specific deposition pressures and powers. Negative thermal expansion coefficients of -3ppm/K have been achieved in air and N_2 from 20 ºC to 300 ºC.
2) Atomic layer deposited(ALD) Al_2O_3 thin films have been show to encapsulate nanoporous thin films. The significance of this innovation is that many thin films that are unstable with changes in ambient often have unusual and desirable optical or mechanical properties (such as low refractive index, low modulus, or negative thermal expansion). These films have not been used in standard optical coatings but not that ALD alumina can be used to stabilize them, their drawbacks are no longer detrimental to the optical/mechanical design process.

VIII. Honors/Awards: (List honors and awards received during the grant/contract period. List lifetime achievement honors such as Nobel Prize, honorary doctorate, and society fellowships prior to this effort.)
1) Nicholas T. Gabriel, 3M Fellowship
2) Martin T. K. Soh, Fulbright Fellowship

IX. Markings: (In order to ensure prompt receipt and acceptance, mark the outside of the package clearly to indicate that it is a performance report.)